

Linear response theory for quantum open systems

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Basing on the theory of Feynman's influence functional and its hierarchical equations of motion, we develop a linear response theory for quantum open systems. Our theory provides an effective way to calculate dynamical observables of a quantum open system at its steady-state, which can be applied to various fields of non-equilibrium condensed matter physics.

Introduction: The linear response theory (LRT) has been widely used in condensed matter physics since it was derived by Kubo in 1957 [1]. For example, one can calculate the conductivity via the current-current correlation function directly from the ground state without applying any realistic bias voltage. However, Kubo's theory is only valid for an equilibrium closed system (not limited to quantum one). In recent years, quantum theory for open systems have attract more and more research interests in molecular electronics, nanophysics and biophysics, etc. One fundamental issue is whether we can directly calculate the dynamical observables (e.g. spectra function) of a static open system (with time-translation-symmetry) instead of evolving it under some time-dependent field? In this letter, we address this issue by developing a linear response theory for quantum open systems (in analogy with Kubo's). Our derivation is based on the theory of Feynman's influence functional [2] and its hierarchical equations of motion (HEOM) [3].

There exist two correlated questions in developing the LRT for quantum open systems, 1) how to get the reduced density operator (RDO) of a static open system; and 2) how to calculate the response to another probe field? Since question 1 has been well resolved recently via the HEOM of RDO (and its auxiliary ones) derived by differentiating Feynman's influence functional [3], we thus focus on question 2 in the present work.

Before the formal derivation, let us introduce an important character of influence functional: if a system S simultaneously interacts with two different environments (A and B) and no direct coupling between them exists at initial conditions, then the total influence functional F of the system is: $F = F_A \cdot F_B$ [2]. It also means the influence phase satisfying: $\Phi(t) = \Phi_A(t) + \Phi_B(t)$ [2]. When one differentiates the influence functional to derive the HEOM [3], those two different phases will linearly enter into HEOM tie by tie with their simple relationship being maintained [see Eq.(5)]. That character provides a practical solution for question 2 mentioned above.

HEOM of propagator: As the answer of question 1 (and for later purposes), we first outline the main results of Ref.3 in the language of Liouville-space propa-

gator. Let us suppose our quantum open system composed of the (reduced) system with particle operator a_μ^σ ($\sigma = +/-$ corresponding to the creation/annihilation operator) and the environment with bath correlation function $C_{\mu\nu}^\sigma(t)$ (For fermion bath, $\sigma = +/-$ corresponding to particle transferring in/out of the system), where μ (ν) represents the orbital (site, energy or spin, etc.) index of the system. We then chose $\{\psi\}$ as an arbitrary basis set (defining a certain path in path integral representation) in the system subspace, and $\psi \equiv \{\psi, \psi'\}$ for short of the two paths in the theory of influence functional [2]. Now we can write the HEOM of the influence functional in Liouville space as $[j = \{\mu\sigma\}, \bar{j} = \{\mu\bar{\sigma}\}]$ [4][5]

$$\partial_t \mathcal{F}_j^{(n)} = \tilde{\mathcal{F}}_j^{(n)} - i \sum_{k=1}^n (-1)^{n-k} \tilde{\mathcal{C}}_{j_k} \mathcal{F}_{j_k}^{(n-1)} - i \sum_j' \mathcal{A}_j \mathcal{F}_{j\bar{j}}^{(n+1)}, \quad (1)$$

where the sum \sum' runs over all $j \neq j_k$; $k = 1, \dots, n$. $\mathcal{F}^{(0)}$ is the superoperator form of the ordinary influence functional F , and $\mathcal{F}^{(n \geq 1)}$ the auxiliary ones. The expressions of auxiliary influence functionals appearing in the rhs of Eq.(1) are [6]

$$\tilde{\mathcal{F}}_j^{(n)} \equiv (\tilde{\mathcal{B}}_{j_n} \mathcal{B}_{j_{n-1}} \cdots \mathcal{B}_{j_1} + \cdots + \mathcal{B}_{j_n} \cdots \mathcal{B}_{j_2} \tilde{\mathcal{B}}_{j_1}) \mathcal{F} \quad (2a)$$

$$\mathcal{F}_{j_k}^{(n-1)} \equiv \mathcal{B}_{j_n} \cdots \mathcal{B}_{j_{k+1}} \mathcal{B}_{j_{k-1}} \cdots \mathcal{B}_{j_1} \mathcal{F}; \quad (2b)$$

$$\mathcal{F}_{j\bar{j}}^{(n+1)} \equiv \mathcal{B}_j \mathcal{B}_{j_n} \cdots \mathcal{B}_{j_1} \mathcal{F}. \quad (2c)$$

The auxiliary RDOs, $\rho_j^{(n)}$, can hen be defined in terms of the auxiliary influence functionals

$$\rho_j^{(n)}(t) \equiv \mathcal{U}_j^{(n)}(t, t_0) \rho(t_0), \quad (3)$$

where $\mathcal{U}_j^{(n)}(t, t_0)$ is the time-evolution superoperator

$$\mathcal{U}_j^{(n)}(\psi, t; \psi_0, t_0) \equiv \int_{\psi_0[t_0]}^{\psi[t]} \mathcal{D}\psi e^{iS[\psi]} \mathcal{F}_j^{(n)}[\psi] e^{-iS[\psi']}, \quad (4)$$

with $S[\psi]$ being the classical action functional of the system.

From Eq.(1), one can derive the HEOM of RDO as shown in Ref.3. Here, we give the HEOM of the reduced

Liouville-space propagator, defined as $\mathcal{G}_{\mathbf{j}}^{(n)}(t - t_0) \equiv \mathcal{U}_{\mathbf{j}}^{(n)}(t, t_0)$ for the time-translation-invariant system

$$\begin{aligned} \dot{\mathcal{G}}(t) &= -[i\mathcal{L}_s + i\mathcal{L}_{sf}(t)]\mathcal{G}(t) - i\sum_j \mathcal{A}_{\bar{j}}\mathcal{G}_{\mathbf{j}}^{(1)}(t); \quad (5a) \\ \dot{\mathcal{G}}_{\mathbf{j}}^{(n)}(t) &= -[i\mathcal{L}_s + i\mathcal{L}_{sf}(t)]\mathcal{G}_{\mathbf{j}}^{(n)}(t) + \tilde{\mathcal{G}}_{\mathbf{j}}^{(n)}(t) - i\sum_{k=1}^n \\ &\quad \times (-1)^{n-k} \tilde{\mathcal{C}}_{j_k} \mathcal{G}_{\mathbf{j}_k}^{(n-1)}(t) - i\sum_j {}'\mathcal{A}_{\bar{j}}\mathcal{G}_{\mathbf{j}\bar{j}}^{(n+1)}(t). \quad (5b) \end{aligned}$$

with the initial condition being $\mathcal{G}_{\mathbf{j}}^{(n)}(t = 0) = \delta_{n0}$. In Eq.5, \mathcal{L}_s is the Liouville operator of the system, $\mathcal{L}_s \dots \equiv [H_s, \dots]$, while $\mathcal{L}_{sf}(t)$ is that of arbitrary external time-dependent field (e.g. the probe field in the LRT).

HEOM space : We now define the HEOM linear space that can be seen as an extension of Liouville space. In the former the basic element is no longer a operators as in the latter, but is $(N + 1)$ dimensional super-vector constituted by the operator and its auxiliary ones. Let us take the RDO as an example, which is extended as the HEOM-space RDO, i.e.

$$\boldsymbol{\rho}(t) \equiv \{\rho(t), \rho_{\mathbf{j}}^{(n)}(t) [n = 1, 2 \dots N]\}. \quad (6)$$

According to Eqs.(3) and (5), the time-evolution of $\boldsymbol{\rho}(t)$ is determined by the HEOM-space reduced propagator $\hat{\mathcal{G}}(t, t_0)$

$$\boldsymbol{\rho}(t) = \hat{\mathcal{G}}(t, t_0)\boldsymbol{\rho}(t_0). \quad (7)$$

In HEOM space, Eq.(5) can be shorted as

$$\partial \hat{\mathcal{G}}(t, t_0) / \partial t = -\hat{\mathbf{A}}(t)\hat{\mathcal{G}}(t, t_0), \quad (8)$$

where $\hat{\mathbf{A}}(t)$ is the main time-evolution superoperator acting on super-vectors in HEOM space, which determines the equation of motion (EOM) of the propagator as well as the density operator

$$\dot{\boldsymbol{\rho}}(t) = -\hat{\mathbf{A}}(t)\boldsymbol{\rho}(t). \quad (9)$$

Although the concrete form of $\hat{\mathbf{A}}(t)$ can not be written out separately from Eq.(8), one can easily work out its effect from Eq.(5)

Generalizing above definitions, any operator in Hilbert space now can be expanded to a super-vector in HEOM space, which obeys similar EOM as Eq.(9),

$$\mathbf{A}(t) \equiv \{A, A_{\mathbf{j}}^{(n)} (n = 1, 2 \dots N)\}. \quad (10)$$

The inner product of two vectors \mathbf{A} and \mathbf{B} in HEOM space is defined as

$$\langle\langle \mathbf{A} | \mathbf{B} \rangle\rangle \equiv \langle\langle A | B \rangle\rangle + \sum_{n=1}^N \langle\langle A_{\mathbf{j}}^{(n)} | B_{\mathbf{j}}^{(n)} \rangle\rangle, \quad (11)$$

where $\langle\langle A | B \rangle\rangle \equiv \text{Tr}[A^+ B]$ being the standard definition of inner product in Liouville space. Similarly, $\langle\langle A_{\mathbf{j}}^{(n)} | B_{\mathbf{j}}^{(n)} \rangle\rangle \equiv \text{Tr}\left\{[A_{\mathbf{j}}^{(n)}]^+ B_{\mathbf{j}}^{(n)}\right\}$.

LRT in HEOM space: We are now on the position to develop the linear response theory in HEOM space. Suppose a variable A of the system S is perturbed by a weak probe field $\epsilon_{pr}(t)$, which linearly couples to the system via its another variable B

$$H_{pr}(t) = -B\epsilon_{pr}(t). \quad (12)$$

For simplicity, we suppose both A and B are Hermitian operators (extending to non-Hermitian cases is straightforward). The change of the expected value of A caused by the disturbance of $\epsilon_{pr}(t)$ is

$$\delta \bar{A}(t) = \text{Tr}[A \delta \boldsymbol{\rho}(t)] = \langle\langle \mathbf{A}(0) | \delta \boldsymbol{\rho}(t) \rangle\rangle, \quad (13)$$

where $\mathbf{A}(0)$ and $\delta \boldsymbol{\rho}(t)$ respectively denote the expansion of A and $\delta \rho(t)$ in HEOM space, i.e.

$$\mathbf{A}(0) = \{A, \mathbf{0}\}; \quad (14a)$$

$$\delta \boldsymbol{\rho}(t) \equiv \{\delta \rho(t), \delta \rho_{\mathbf{j}}^{(n)}(n = 1, 2 \dots N)\}. \quad (14b)$$

We then apply the first order perturbation in the EOM of RDO,

$$\boldsymbol{\rho}(t) = \boldsymbol{\rho}_s(t) + \delta \boldsymbol{\rho}(t) = \hat{\mathcal{G}}(t, \tau)\boldsymbol{\rho}(\tau). \quad (15)$$

By separating the superoperator $\hat{\mathbf{A}}(t)$ as $\hat{\mathbf{A}}(t) = \hat{\mathbf{A}}_s + \hat{\mathbf{A}}_{pr}(t)$, and inserting the Dyson's equation in HEOM space[7] into Eq.15

$$\hat{\mathcal{G}}(t, \tau) = \hat{\mathcal{G}}_s(t - \tau) - \int_{\tau}^t d\tau' \hat{\mathcal{G}}(t, \tau') \hat{\mathbf{A}}_{pr}(\tau') \hat{\mathcal{G}}_s(\tau' - \tau), \quad (16)$$

we arrive at

$$\delta \boldsymbol{\rho}(t) = - \int_0^t d\tau \hat{\mathcal{G}}(t, \tau) \hat{\mathbf{A}}_{pr}(\tau) \boldsymbol{\rho}_s(\tau). \quad (17)$$

In above derivation, we have used $\boldsymbol{\rho}_s(t) = \hat{\mathcal{G}}_s(t - \tau)\boldsymbol{\rho}_s(\tau)$ and initial conditions $\delta \boldsymbol{\rho}(\tau = 0) = 0$; $\boldsymbol{\rho}(\tau = 0) = \boldsymbol{\rho}_s(\tau = 0)$.

To proceed, we define the time-independent HEOM-space superoperator $\hat{\mathbf{B}}$ as

$$\hat{\mathbf{B}} \equiv i\hat{\mathbf{A}}_{pr}(t)/\epsilon_{pr}(t), \quad (18)$$

whose action can be determined from $\hat{\mathbf{A}}_{pr}(t)\mathbf{A} = i[H_{pr}(t), \mathbf{A}]$ [see Eq.(5)] as

$$\hat{\mathbf{B}}\rho = [B, \rho]. \quad (19)$$

Inserting Eq.(19) into (17), we finally get

$$\delta\bar{A}(t) = i \int_0^t d\tau \left\langle \left\langle \mathbf{A}(0) | \hat{\mathbf{G}}(t, \tau) \hat{\mathbf{B}} | \rho(\tau) \right\rangle \right\rangle \epsilon_{pr}(\tau), \quad (20)$$

from which we are ready to define the response function in HEOM space as

$$\chi_{AB}(t, \tau) \equiv i \left\langle \left\langle \mathbf{A}(0) | \hat{\mathbf{G}}(t, \tau) \hat{\mathbf{B}} | \rho(\tau) \right\rangle \right\rangle. \quad (21)$$

Obviously, the definition of Eq. (21) is quite general. For a static quantum open system satisfying time-translation-symmetry, $\rho(\tau) \rightarrow \rho_{eq}(T) \equiv \{\rho_{eq}, \rho_{eq}^{(n)}[n = 1, 2, \dots, N]\}$ and $\hat{\mathbf{G}}(t, \tau) \rightarrow \hat{\mathbf{G}}_s(t - \tau)$, which leads to following definition of the response function

$$\chi_{AB}(t) \equiv i \left\langle \left\langle \mathbf{A}(0) | \hat{\mathbf{G}}_s(t) \hat{\mathbf{B}} | \rho_{eq}(T) \right\rangle \right\rangle. \quad (22)$$

The correlation function in HEOM space can be similarly defined as [8]

$$\tilde{C}_{AB}(t) \equiv \left\langle \left\langle \mathbf{A}(0) | \hat{\mathbf{G}}_s(t) \mathbf{B} | \rho_{eq}(T) \right\rangle \right\rangle, \quad (23)$$

where $\mathbf{B}\rho_{eq}(T) = \{B\rho_{eq}, B\rho_{eq}^{(n)}(n = 1, 2, \dots, N)\}$.

For practical calculation, we can rewrite Eq.(22) as

$$\chi_{AB}(t) = \langle \langle \mathbf{A}(0) | \sigma(t) \rangle \rangle = \text{Tr} [A^+ \sigma(t)], \quad (24)$$

where

$$\sigma(t) = \hat{\mathbf{G}}_s(t)\sigma(0); \quad (25a)$$

$$\begin{aligned} \sigma(0) &= i\hat{\mathbf{B}}\rho_{eq}(T) \\ &= \{i[B, \rho_{eq}], i[B, \rho_{eq}^{(n)}](n = 1, 2, \dots, N)\}. \end{aligned} \quad (25b)$$

Similarly, if we set $\sigma(0) = \mathbf{B}\rho_{eq}(T)$, Eq.(23) can be rewritten as an easier handling form

$$\tilde{C}_{AB}(t) = \langle \langle \mathbf{A}(0) | \sigma(t) \rangle \rangle = \text{Tr} [A^+ \sigma(t)]. \quad (26)$$

Spectra function in HEOM space: In principle, one can calculate the dynamical correlation of any two system-operators (A and B) via the HEOM-space LRT. One typical example is the spectra function that plays important role in many body physics. In what follows, we will demonstrate how to obtain the (reduced) spectra function of a quantum open system. The spectra function $J_{AB}(\omega)$ directly relates to the imaginary part of the retarded single-particle Green's function $G_{AB}^r(t)$ in the form of

$$G_{AB}^r(t) \equiv -i\theta(t) \langle \{A(t), B\} \rangle; \quad (27a)$$

$$J_{AB}(\omega) \equiv -\frac{1}{\pi} \text{Im} [G_{AB}^r(\omega)]. \quad (27b)$$

Please be noted that the Green's function in our theory is defined for two arbitrary operators, which can reduce to the one in textbooks by setting $A = a$ and $B = a^+$.

For fermion, $G_{AB}^r(t)$ can not be obtained directly from $\chi_{AB}(t)$ due to the anti-communication relation in the former but communication one in the latter. Fortunately, we can get $G_{AB}^r(t)$ from correlation function $\tilde{C}_{AB}(t)$, i.e.

$$\begin{aligned} G_{AB}^r(t) &= -i\theta(t) \langle \{A(t), B\} \rangle \\ &= -i\theta(t) [\tilde{C}_{AB}(t) + \tilde{C}_{BA}(-t)]. \end{aligned} \quad (28)$$

To proceed, we introduce the general spectra function $C_{AB}(\omega) \equiv \frac{1}{2} \int_{-\infty}^{+\infty} dt e^{i\omega t} \tilde{C}_{AB}(t)$, which satisfies the detailed-balance-relation $C_{BA}(-\omega) = e^{-\beta\omega} C_{AB}(\omega)$. After some algebra, we obtain

$$J_{AB}(\omega) = \frac{1}{\pi} (1 + e^{-\beta\omega}) C_{AB}(\omega), \quad (29)$$

which is obviously the fluctuation-dissipation theorem in HEOM space.

Since in Eq.(23) only $\hat{\mathbf{G}}_s(t)$ is the function of time, we have

$$C_{AB}(\omega) = \left\langle \left\langle \mathbf{A}(0) | \hat{\mathbf{G}}_s(\omega) \mathbf{B} | \rho_{eq}(T) \right\rangle \right\rangle, \quad (30)$$

where $\hat{\mathbf{G}}_s(\omega)$ is the Fourier transform of Eq.(5). Thus $J_{AB}(\omega)$ can be calculated from

$$J_{AB}(\omega) = \frac{1}{2\pi} (1 + e^{-\beta\omega}) \left\langle \left\langle \mathbf{A}(0) | \hat{\mathbf{G}}_s(\omega) \mathbf{B} | \rho_{eq}(T) \right\rangle \right\rangle. \quad (31)$$

More concretely, if choosing $\sigma(0) = \mathbf{B}\rho_{eq}(T) = \{B\rho_{eq}, B\rho_{eq}^{(n)}(n = 1, 2, \dots, N)\}$, then we have $\sigma(\omega) = \hat{\mathbf{G}}_s(\omega)\sigma(0)$ and

$$\begin{aligned} J_{AB}(\omega) &= \frac{1}{2\pi} (1 + e^{-\beta\omega}) \langle \langle \mathbf{A}(0) | \sigma(\omega) \rangle \rangle \\ &= \frac{1}{2\pi} (1 + e^{-\beta\omega}) \text{Tr} [A^+ \sigma(\omega)]. \end{aligned} \quad (32)$$

Eq.(32) is the main formula to calculate HEOM-space spectra function in our theory. Before that, one must solve equation $\hat{\mathbf{A}}_s \boldsymbol{\rho}_{eq}(T) = 0$ to obtain $\boldsymbol{\rho}_{eq}(T)$.

Summary: In summary, we have developed a linear response theory for quantum open systems on the basis of the theory of Feynman's influence functional and its hierarchical equations of motion. From our theory, one can directly calculate the dynamical observables (e.g. spectra function) of a static open system instead of evolving it under time-dependent field. It can be applied to non-equilibrium many-body physics, nanophysics, etc.

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$$\mathcal{A}_j \equiv a_\mu^\sigma[\psi(t)] + a_\mu^\sigma[\psi'(t)].$$

[5]

$$\tilde{\mathcal{C}}_j \equiv \sum_\nu C_{\mu\nu}^\sigma(t=0) a_\nu^\sigma[\psi(t)] - \sum_\nu C_{\nu\mu}^{\bar{\sigma}}(t=0) a_\nu^\sigma[\psi'(t)].$$

[6] $\mathcal{B}_j = -i \sum_\nu \int_{t_0}^t d\tau C_{\mu\nu}^\sigma(t, \tau) a_\nu^\sigma[\psi(\tau)] + i \sum_\nu \int_{t_0}^t d\tau C_{\mu\nu}^{\bar{\sigma}*}(t, \tau) a_\nu^\sigma[\psi'(\tau)]$. $\tilde{\mathcal{B}}_j$ is similar to \mathcal{B}_j but with $C_{\mu\nu}^\sigma/C_{\mu\nu}^{\bar{\sigma}*}$ replaced by $\dot{C}_{\mu\nu}^\sigma/\dot{C}_{\mu\nu}^{\bar{\sigma}*}$.

[7] The validity of Dyson's equation in HEOM space is natural. One can directly verify Eq.(16) by inserting it into Eq.(5) and letting $\mathcal{L}_{sf}(t) = \mathcal{L}_{pr}(t)$.

[8] In our theory, the fluctuation-dissipation theorem is naturally established. See Eq.(29).